(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) Publication number: **0 308 198**
A2

(12) EUROPEAN PATENT APPLICATION

(21) Application number: 88308496.4

(22) Date of filing: 14.09.88

(51) Int. Cl.4: C 08 L 33/24
C 08 F 283/12, C 08 G 77/20,
C 08 G 77/28
//(C08L33/24,51:08)

(30) Priority: 18.09.87 JP 234050/87

(43) Date of publication of application:
22.03.89 Bulletin 89/12

(84) Designated Contracting States: DE FR GB

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(54) Thermoplastic polymethacrylimide resin composition.

(57) A polymethacrylamide resin composition having an improved impact resistance and thermal resistance is prepared by blending a polymethacrylamide resin with a polyorganosiloxane type graft copolymer formed by graft-polymerizing at least one ethylenically unsaturated monomer onto a polyorganosiloxane rubber copolymerized with a graft-crosslinking agent.

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From the results obtained in the examples and comparative examples, it is seen that the impact resistance and flowability were greatly improved in the resin compositions of the examples. In the resin compositions of Examples 1, 5, 9, 4, 8, and 12, γ-methacryloyloxypropyldimethoxymethylsilane and γ-methacryloyloxypropyltrimethoxysilane were used as the graft-crosslinking agent, and in the resin compositions of Examples 2, 6, 10, 3, 7, and 11, γ-mercaptopropyldimethoxysilane and tetramethyltetravinylcyclosiloxane were used as the graft-crosslinking agent. It is seen that the former resin compositions were superior to the latter compositions in Izod impact strength and melt index. This is due to the difference of the graft efficiency, as when the graft efficiency is high, even if the polymethacrylimide resin is combined with the polyorganosiloxane type graft copolymer, the impact resistance can be improved without inhibiting the flowability.

Examples 13 through 16 and Comparative Example 4

The polymethacrylimide resin A-1 obtained in Referential Example 1 was mixed with the polyorganosiloxane type graft copolymer S-1 obtained in Referential Example 2 at various mixing ratios (Examples 13 through 16).

An MBS resin (a graft copolymer having a butadiene rubber content of 60% and a methyl methacrylate/styrene weight ratio of 25/15) was mixed with the above-mentioned polymethacrylimide resin at a mixing ratio shown in Table 4 (Comparative Example 4).

Each composition was pelletized under the same conditions by using the same twin-screw extruder and injection molding machine as described in the foregoing examples, and the physical properties of the obtained pellet and test piece were evaluated. The results are shown in Table 4.

Table 4

		Amount of Polymethacryl-imide Resin A-1 (%)	Amount of Polyorgano-siloxane Type Graft Copolymer S-1 (%)	MBS Resin (%)	Izod Impact Strength (with 1/4", notch, 23°C) (kg·cm/cm)	Heat Distortion Temperature (°C)	Melt Index (10 kg, 260°C) (g/10 min)	Flexural Strength (kg/cm^2)
Example No.	13	92.7	7.3	-	13	140	13	1350
	14	89.2	10.8	-	17	140	10	1350
	15	87.5	12.5	-	20	139	8	1350
	16	83.3	16.7	-	25	138	7	1350
Comparative Example No.	4	92.7	-	7.3	6.5	138	4	1200

As apparent from the results of Examples 13 through 16 shown in Table 4, the impact strength was increased with an increase of the mixing ratio of the graft copolymer S-1.

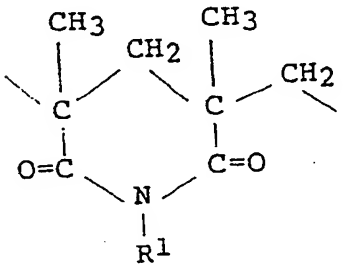
When the compositions of these examples are compared with the composition comprising the MBS resin and the polymethacrylimide resin, it is seen that the latter composition has an inferior impact strength, flowability, and mechanical strength.

According to the present invention, a thermoplastic resin composition having an excellent impact resistance, heat resistance, mechanical strength, moldability, and flowability is obtained by the incorporation of a specific polyorganosiloxane type graft copolymer in a polymethacrylimide resin.

Claims

1. A polymethacrylimide resin composition comprising (A) a polymethacrylimide resin and (B) a polyorganosiloxane type graft copolymer formed by graft-polymerizing at least one ethylenically unsaturated monomer onto a polyorganosiloxane rubber copolymerized with a graft-crosslinking agent.

2. A polymethacrylimide resin composition according to claim 1, wherein the polymethacrylimide resin (A) comprises at least 5% by weight of imide ring structural units represented by the following general formula (1):



(1)

wherein R^1 represents a hydrogen atom, or an alkyl, cycloalkyl, aryl, alkaryl, aralkyl or allyl group having 1 to 20 carbon atoms.

3. A polymethacrylimide resin composition according to claim 1, wherein the polymethacrylimide resin (A) comprises at least 20% by weight of the imide ring structural units.

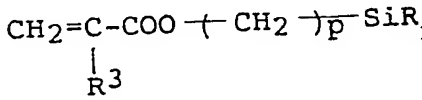
4. A polymethacrylimide resin composition according to claim 1, wherein the polymethacrylimide resin (A) is prepared by reacting a methacrylate resin with ammonia or a primary amine having an alkyl, cycloalkyl, aryl, alkaryl, aralkyl or allyl group having 1 to 20 carbon atoms, at a temperature of 150 to 350° C in an inert solvent.

5. A polymethacrylimide resin composition according to claim 4, wherein the methacrylate resin is a homopolymer of methyl methacrylate or a copolymer comprised of at least 25% by weight of methyl methacrylate and not more than 75% by weight of at least one ethylenically unsaturated monomer copolymerizable therewith.

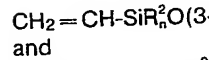
6. A polymethacrylimide resin composition according to claim 1, wherein R^1 in the formula (1) is selected from the group consisting of hydrogen, methyl, ethyl, propyl, butyl, phenyl and cyclohexyl.

7. A polymethacrylimide resin composition according to claim 1, wherein the amount of the polyorganosiloxane rubber is 5 to 90% by weight based on the weight of the polyorganosiloxane type graft copolymer (B).

8. A polymethacrylimide resin composition according to claim 1, wherein the polyorganosiloxane rubber is obtained by polymerizing, based on the weight of the polyorganosiloxane rubber, at least 50% by weight of an organsiloxane with 0.1 to 20% by weight of the graft-crosslinking agent and 0.2 to 30% by weight of a crosslinking agent; said graft-crosslinking agent being selected from the group consisting of organosiloxane compounds represented by the following formulae (2), (3) and (4):

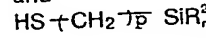


(2),



(3)

and



(4)

wherein R^2 stands for a methyl, ethyl, propyl or phenyl group, R^3 stands for hydrogen or a methyl group, n is 0, 1 or 2 and p is a number of from 1 to 6;
and said crosslinking agent is a trifunctional or tetrafunctional organosiloxane type crosslinking agent.

9. A polymethacrylimide resin composition according to claim 8, wherein the graft-crosslinking agent is an organosiloxane compound represented by the formula (2).

10. A polymethacrylimide resin composition according to claim 1, wherein the polyorganosiloxane rubber has an average particle diameter of 0.06 to 0.6 μm.

11. A polymethacrylimide resin composition according to claim 1, wherein the polyorganosiloxane type graft copolymer is formed by graft-copolymerizing 10 to 95% by weight, based on the weight of the graft copolymer, of at least one ethylenically unsaturated monomer onto 90 to 5% by weight, based on the weight of the graft copolymer, of the polyorganosiloxane rubber.

12. A polymethacrylimide resin composition according to claim 1, which comprises (A) 60 to 91% by weight, based on the weight of the total resin composition, of the polymethacrylimide resin and (B) 1 to 40% by weight, based onthe weight of the total resin composition, of the polyorganosiloxane type graft copolymer.

13. An article moulded from a composition according to any preceding claim.

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(30) Priority: 18.09.87 JP 234050/87

(43) Date of publication of application: 22.03.89 Bulletin 89/12

(84) Designated Contracting States: DE FR GB

(88) Date of deferred publication of the search report: 27.06.90 Bulletin 90/26

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EUROPEAN SEARCH REPORT

Application Number

EP 88 30 8496

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
A	US-A-4 246 374 (KOPCHIK) * Claims 24-26 * -----	1	C 08 L 33/24 C 08 F 283/12 C 08 G 77/20 C 08 G 77/28 // (C 08 L 33/24 C 08 L 51:08)

TECHNICAL FIELDS SEARCHED (Int. Cl.4)

C 08 L
C 08 F

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	15-02-1990	SCHUELER D.H.H.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
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E : earlier patent document, but published on, or after the filing date
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